Diffusiophoresis of a Spherical Volatile Particle

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Abstract. A spherical condensed phase (volatile particle) is placed in an infinite expanse of a binary mixture consisting of the vapor of the condensed phase and another noncondensable gas. The mixture is assumed to be at rest with uniform pressure and temperature at infinity. However, the vapor and the noncondensable gas have small uniform density (or concentration) gradients of the same magnitude but in the opposite direction. The flows of both components around the particle are analyzed numerically by a finite-difference method on the basis of a model Boltzmann equation for gas mixtures, and the force acting on the particle (diffusiophoretic force) is obtained accurately for a wide range of the Knudsen number.

INTRODUCTION

Thermophoresis, photophoresis, and diffusiophoresis are typical phenomena due to the effect of gas rarefaction that manifest themselves in connection with aerosol particles and have been important subjects in kinetic theory of gases [1, 2, 3]. Recent advances in microtechnology have reinforced their importance. Among these phenomena, diffusiophoresis [4, 5, 6, 7, 8], the force acting on an aerosol particle and its resultant motion in a mutually diffusing background mixture of gases, is peculiar to gas mixtures and thus has more nonintuitive features.

In the present study, we investigate the diffusiophoresis of a volatile particle. To be more specific, we consider a spherical condensed phase placed in a binary mixture at rest consisting of the vapor of the condensed phase and another noncondensable gas, each of which has a small density (or concentration) gradient of the same magnitude but in the opposite direction. In this situation, evaporation and condensation of the vapor taking place on the surface of the particle may have an important effect on the flows of both components around the particle. We will analyze such flows for a wide range of the Knudsen number numerically on the basis of the linearized version of a model Boltzmann equation [9, 10] and obtain the force acting on the particle (diffusiophoretic force) accurately.

FORMULATION OF THE PROBLEM

Problem and assumptions

Let us consider an infinite expanse of a binary mixture of two gases, gas A and gas B. There is no macroscopic motion of the total mixture, and its temperature and pressure (and thus its molecular number density) are uniform. But, the molecular number densities of respective gases have uniform gradients with the same magnitude but in the opposite directions. Let us denote the uniform temperature by T_0 , the uniform pressure by p_0 , and the uniform molecular number density by $n_0 = p_0/\kappa T_0$, where κ is the Boltzmann constant), and let us express the number density of gas A as $n_0^A + (\partial n^A/\partial X_1)_{\infty}X_1$ and that of gas B as $n_0^B - (\partial n^A/\partial X_1)_{\infty}X_1$, where $n_0^A + n_0^B = n_0$, and $n_0^A + n_0^A = n_0^A$, and that of gas $n_0^A + n_0^A = n_0^A + n_0^A = n_0^A + n_0^A + n_0^A = n_0^A + n_0^A + n_0^A = n_0^A + n_0^A + n_0^A + n_0^A = n_0^A + n_0^A +$

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Form Approved OMB No. 0704-0188 number on the basis of kinetic theory under the following assumptions:

- (i) The behavior of the mixture is described by the model Boltzmann equation proposed by Hamel [9] or that by Garzó *et al.* [10]. [Once the linearization around an equilibrium state at rest is made, these two models give the same equation. We will actually handle the linearized equation, as mentioned in (v) below.]
- (ii) The molecules of the vapor leaving the surface of the particle are distributed according to the Maxwellian distribution corresponding to the vapor in the saturated equilibrium state at rest with temperature T_0 .
- (iii) The molecules of the noncondensable gas undergo diffuse reflection on the particle surface.
- (iv) The n_0^A is equal to the molecular number density of the vapor in the saturated equilibrium state mentioned in (ii).
- (v) The imposed gradient of the number density (or the concentration) of each gas is small $[|(\partial n^A/\partial X_1)_{\infty}| \ll n_0^A/L$, or $|(\partial \chi^A/\partial X_1)_{\infty}| \ll \chi_0^A/L]$, so that the equation and boundary condition can be linearized.

The assumption (iv) as well as the assumption that the temperature of the particle is the same as that of the mixture at infinity (T_0) is not essential to the present result, as will be explained in "Results and discussions". The case where there is a uniform flow of the mixture at infinity will also be discussed there.

Basic equations

We first summarize the main notations. In what follows, the Greek letters α and β stand for the labels A and B of the component gases. For gas α ($\alpha=A$, B), F^{α} is the velocity distribution function, n^{α} the molecular number density, v_i^{α} the flow velocity, T^{α} the temperature, and m^{α} the mass of a molecule. Our reference state is the equilibrium state at rest with temperature T_0 and number densities of respective gases n_0^A and n_0^B , which is expressed as $F_0^{\alpha} = n_0^{\alpha} (2\kappa T_0/m^A)^{-3/2} E^{\alpha}$ with $E^{\alpha} = (\hat{m}^{\alpha}/\pi)^{3/2} \exp(-\hat{m}^{\alpha}\zeta_i^2)$, where $\hat{m}^{\alpha} = m^{\alpha}/m^A$ and ζ_i is the dimensionless molecular velocity $[(2\kappa T_0/m^A)^{1/2}\zeta_i$ is the dimensional one]. Then, we let $F^{\alpha} = F_0^{\alpha}(1+\phi^{\alpha})$, $n^{\alpha} = n_0^{\alpha}(1+\omega^{\alpha})$, $v_i^{\alpha} = (2\kappa T_0/m^A)^{1/2}u_i^{\alpha}$, $T^{\alpha} = T_0(1+\tau^{\alpha})$, so that $|\phi^{\alpha}|$, $|\omega^{\alpha}|$, $|u_i^{\alpha}|$, $|\tau^{\alpha}| \ll 1$. In addition, $x_i = X_i/L$ and $\hat{n}_0^{\alpha} = n_0^{\alpha}/n_0^A$. For the model equation, the collision frequency of the α -gas molecules for the collisions with the β -gas molecules is given by $K^{\alpha\beta}n^{\beta}$ with constants $K^{\alpha\beta}$ ($K^{AB} = K^{BA}$). Then, the mean free path of the vapor molecules ℓ_0^A in the reference equilibrium state is given by $\ell_0^A = (8\kappa T_0/\pi m^A)^{1/2}(K^{AA}n_0^A + K^{AB}n_0^B)^{-1}$. With $K^{\alpha\beta}$ and ℓ_0^A , we define $\hat{K}^{\alpha\beta} = K^{\alpha\beta}/K^{AA}$ and $k = (\sqrt{\pi}/2)(k_0^A/L)$, where $K^{\alpha\beta}n^A$ is the Knudsen number.

With the above notations, the linearized model equation is written as

$$\zeta_{i} \frac{\partial \phi^{\alpha}}{\partial x_{i}} = \frac{1}{(1 + \hat{K}^{AB} \hat{n}_{0}^{B})k} \sum_{\beta = A} \hat{K}^{\alpha\beta} \hat{n}_{0}^{\beta} (\Psi^{\alpha\beta} - \phi^{\alpha}), \qquad (\alpha = A, B),$$

$$(1)$$

$$\Psi^{\alpha\beta} = \omega^{\alpha} + 2\hat{m}^{\alpha}\zeta_{i}u_{i}^{\alpha\beta} + (\hat{m}^{\alpha}\zeta_{i}^{2} - 3/2)\tau^{\alpha\beta}, \tag{2}$$

$$u_i^{\alpha\beta} = \frac{\hat{m}^{\alpha} u_i^{\alpha} + \hat{m}^{\beta} u_i^{\beta}}{\hat{m}^{\alpha} + \hat{m}^{\beta}}, \qquad \tau^{\alpha\beta} = \tau^{\alpha} + \frac{2\hat{m}^{\alpha} \hat{m}^{\beta}}{(\hat{m}^{\alpha} + \hat{m}^{\beta})^2} (\tau^{\beta} - \tau^{\alpha}), \tag{3}$$

$$\omega^{\alpha} = \int \phi^{\alpha} E^{\alpha} d^{3} \zeta, \quad u_{i}^{\alpha} = \int \zeta_{i} \phi^{\alpha} E^{\alpha} d^{3} \zeta, \quad \tau^{\alpha} = (2/3) \int (\hat{m}^{\alpha} \zeta_{i}^{2} - 3/2) \phi^{\alpha} E^{\alpha} d^{3} \zeta, \tag{4}$$

where $d^3\zeta = d\zeta_1 d\zeta_2 d\zeta_3$. The boundary condition on the surface of the particle ($|x_i| = 1$) is given as

$$\phi^{A} = 0, \qquad \phi^{B} = -2(\pi \hat{m}^{B})^{1/2} \int_{\zeta_{i} n_{i} < 0} \zeta_{j} n_{j} \phi^{B} E^{B} d^{3} \zeta, \quad (\zeta_{i} n_{i} > 0), \tag{5}$$

where n_i is the unit normal vector to the particle surface pointing into the gas. On the other hand, the boundary condition at infinity $(|x_i| \to \infty)$ is given by

$$\phi^{A} \to \phi_{\infty}^{A} = c_{\infty}(x_{1} - 2\zeta_{1}\widetilde{D}k), \qquad \phi^{B} \to \phi_{\infty}^{B} = -\frac{c_{\infty}}{\hat{n}_{0}^{B}}(x_{1} - 2\zeta_{1}\widetilde{D}k), \qquad \widetilde{D} = \frac{(1 + \hat{K}^{AB}\hat{n}_{0}^{B})(1 + \hat{m}^{B})}{2\hat{K}^{AB}(1 + \hat{m}^{B}\hat{n}_{0}^{B})}, \tag{6}$$

where $c_{\infty} = (L/\chi_0^A)(\partial \chi^A/\partial X_1)_{\infty} = (L/n_0^A)(\partial n^A/\partial X_1)_{\infty}$, and ϕ_{∞}^{α} is the solution to Eq. (1) expressing the situation described in the first paragraph in "Problem and assumptions". That is, this solution gives the following macroscopic variables:

$$\omega^{A} = c_{\infty} x_{1}, \quad \omega^{B} = -c_{\infty} x_{1} / \hat{n}_{0}^{B}, \quad \tau^{\alpha} = 0, \quad u_{1}^{A} = -c_{\infty} \widetilde{D} k, \quad u_{1}^{B} = c_{\infty} \widetilde{D} k / \hat{m}^{B} \hat{n}_{0}^{B}, \quad u_{2}^{\alpha} = u_{3}^{\alpha} = 0.$$
 (7)

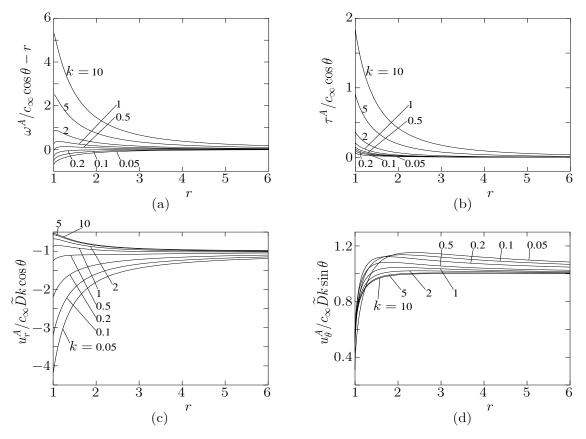


FIGURE 1. Profiles of the macroscopic quantities of the vapor for various values of k in the case where $m^A = m^B$, $K^{AA} = K^{AB} = K^{BB}$, and $n_0^A = n_0^B$. (a) ω^A , (b) τ^A , (c) u_r^A , (d) u_θ^A .

Note that there is a flow of each gas (i.e., diffusion) in the direction opposite to its density or concentration gradient though there is no flow of the total mixture $[u_1 = (u_1^A + \hat{m}^B \hat{n}_0^B u_1^B)/(1 + \hat{m}^B \hat{n}_0^B) = 0$, where $(2\kappa T_0/m^A)^{1/2}u_i$ is the flow velocity of the total mixture].

It is seen from Eqs. (1)–(6) that we need to specify the following five dimensionless parameters: \hat{n}^B , \hat{K}^{AB} , \hat{K}^{BB} , \hat{n}_0^B , k to solve the problem (we do not need to specify c_{∞} because of the linearity of the problem).

NUMERICAL ANALYSIS

In the case of a single-component gas, spherical geometry allows a convenient similarity solution to the linearized Boltzmann equation [11, 12]. In the present problem, the similarity solution of the same form, i.e.,

$$\phi^{\alpha} = \Phi_c^{\alpha}(r, \zeta, \theta_{\zeta})\cos\theta + \zeta_{\theta}\Phi_s^{\alpha}(r, \zeta, \theta_{\zeta})\sin\theta, \tag{8}$$

is shown to be compatible with Eqs. (1)–(6), where (r,θ,φ) is the spherical coordinate system with its origin at the center of the particle and its polar axis along the x_1 axis, ζ_r , ζ_θ , and ζ_φ are, respectively, the r, θ , and φ components of ζ_i , $\zeta = (\zeta_i^2)^{1/2}$, and θ_ζ is the angle between ζ_i and the r direction $[\theta_\zeta = \cos^{-1}(\zeta_r/\zeta), 0 \le \theta_\zeta \le \pi]$. Thus, the independent variables are reduced to r, ζ , and θ_ζ . Correspondingly, $\omega^\alpha/\cos\theta$, $u_r^\alpha/\cos\theta$, $u_\theta^\alpha/\sin\theta$, and $\tau^\alpha/\cos\theta$ turn out to be functions of r only, where u_r^α , u_θ^α , and u_φ^α (=0) are the r, θ , and φ components of u_i^α . We solve the boundary value problem for Φ_c^α and Φ_s^α numerically by a finite-difference method. The method

We solve the boundary value problem for Φ_c^{α} and Φ_s^{α} numerically by a finite-difference method. The method is basically the same as that used for a single-component gas in Ref. [12] where a slow rarefied gas flow past a sphere is analyzed, except that we are handling a simpler model collision term here rather than the original Boltzmann collision integral. The difficulties that we still share with Ref. [12] are: (a) the solution approaches the state at infinity

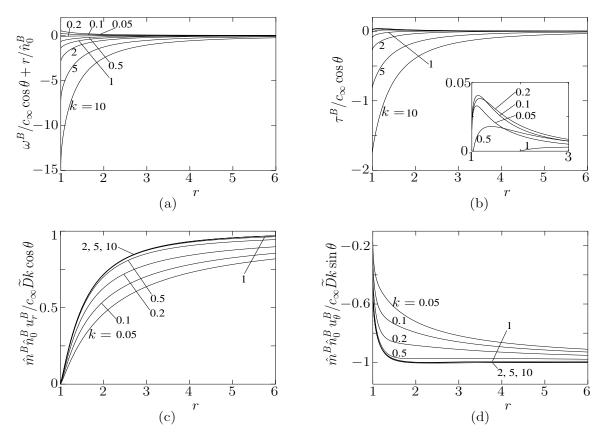


FIGURE 2. Profiles of the macroscopic quantities of the noncondensable gas for various values of k in the case where $m^A = m^B$, $K^{AA} = K^{AB} = K^{BB}$, and $R^A_0 = R^B_0$. (a) ω^B , (b) τ^B , (c) u^B_r , (d) u^B_θ .

very slowly; (b) the velocity distribution functions exhibit discontinuities around the sphere. These are overcome by essentially the same way as in Ref. [12]. That is, (a) is solved by connecting the numerical solution with the explicit asymptotic solution for the far field. The discontinuities in (b) are described accurately by a combination of a standard finite-difference method and a method of characteristics that was devised in Ref. [13].

As a reference, we give the data about the computational systems that are used. For $\hat{m}^B = 0.5$, the computational domain is $1 \le r \le r_M$ [$r_M = 200k$ (k < 5), 750 (k = 5), and 1000 (k = 10)], $0 \le \zeta \le 8$, and $0 \le \theta_\zeta \le \pi$, and the numbers of the grid points in r, ζ , and θ_ζ are 601 (nonuniform), 71 (nonuniform), and 401 (uniform), respectively. The systems for other \hat{m}^B are the same except that $0 \le \zeta \le 7$ and 61 grid points (nonuniform) are used for ζ .

RESULTS AND DISCUSSIONS

First we show the profiles of the macroscopic quantities. Figure 1 shows the profiles for the vapor, that is, $\omega^A/c_\infty\cos\theta-r$, $\tau^A/c_\infty\cos\theta$, $u_r^A/c_\infty\widetilde{D}k\cos\theta$, and $u_\theta^A/c_\infty\widetilde{D}k\sin\theta$ (note that $u_1^A\to -c_\infty\widetilde{D}k$ as $r\to\infty$) versus r, for various k in the case where $m^A=m^B$, $K^{AA}=K^{AB}=K^{BB}$, and $n_0^A=n_0^B$ (or $\hat{m}^B=1$, $\hat{K}^{AB}=\hat{K}^{BB}=1$, and $\hat{n}_0^B=1$). Figure 2 shows the corresponding profiles of the noncondensable gas, that is, $\omega^B/c_\infty\cos\theta+r/\hat{n}_0^B$, $\tau^B/c_\infty\cos\theta$, $\hat{m}^B\hat{n}_0^Bu_r^B/c_\infty\widetilde{D}k\cos\theta$, and $\hat{m}^B\hat{n}_0^Bu_\theta^B/c_\infty\widetilde{D}k\sin\theta$ (note that $u_1^B\to c_\infty\widetilde{D}k/\hat{m}^B\hat{n}_0^B$ as $r\to\infty$) versus r, in the same case. Let us suppose that $c_\infty>0$. Then, there is a uniform flow of gas B in the B in the case of a uniform (noncondensable) gas flow past a sphere [12]. On the other hand, there is a uniform flow of gas A in the negative B in the case of a uniform vapor

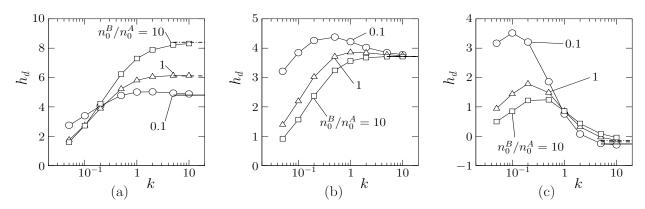


FIGURE 3. The h_d versus k in the case where $K^{AA} = K^{AB} = K^{BB}$. (a) $m^B/m^A = 0.5$, (b) $m^B/m^A = 1$, (c) $m^B/m^A = 2$. The result for the free-molecular flow is shown by a solid line $(n_0^B/n_0^A = 0.1)$, a dashed lone $(n_0^B/n_0^A = 1)$, and a dot-dashed line $(n_0^B/n_0^A = 10)$.

flow past a spherical condensed phase [14]. Thus, condensation takes place on the head (the part facing to the positive x_1 direction) of the particle and evaporation on the tail (the part facing to the negative x_1 direction) [see Fig. 1(c)]. The evaporation and condensation are enhanced by the imposed density gradient c_{∞} that makes the number density around the head higher than the saturation number density of the vapor (at the particle temperature) and that around the tail lower than the saturation number density. The flow field around the sphere is roughly a superposition of these effects.

Next, we show the result of the force acting on the particle (diffusiophoretic force). The force $F_i^d = (F_1^d, 0, 0)$ can be expressed in the following form:

$$F_1^d = \left(\frac{2\kappa T_0}{m^A}\right)^{-1/2} p_0 L^2 D_{AB} \left(\frac{\partial \chi^A}{\partial X_1}\right)_{\infty} h_d, \qquad D_{AB} = \frac{(m^A + m^B)\kappa T_0}{m^A m^B K^{AB} n_0}, \tag{9}$$

where D_{AB} is the mutual diffusion coefficient for the present model Boltzmann equation. The h_d versus k is shown in Fig. 3 in the case where $K^{AA} = K^{AB} = K^{BB} = 1$. Figure 3(a) is for $m^B/m^A = 0.5$, Fig. 3(b) for $m^B/m^A = 1$, and Fig. 3(c) for $m^B/m^A = 2$. In each figure, the results for $n_0^B/n_0^A = 0.1$, 1, and 10 are shown. The h_d is positive in most cases. However, it is rather sensitive to the molecular mass ratio m^B/m^A and becomes negative when $m^B/m^A = 2$ and k is relatively large. The result for the free-molecular flow $(k = \infty)$:

$$h_d = (4\sqrt{\pi}/3)\sqrt{\hat{m}^B}\widetilde{D}\left[8(1-\sqrt{\hat{m}^B}) + \pi\right]/(1+\hat{m}^B),\tag{10}$$

where \widetilde{D} is given in Eq. (6), is also shown in Fig. 3.

In the present study, we assumed that the temperature of the particle, say T_w , was the same as that of the gas at infinity (T_0) and that the reference number density of the vapor (n_0^A) was equal to the saturation number density, say n_s^A , of the vapor at temperature T_w (= T_0). If these conditions are not satisfied (T_w is still assumed to be uniform), but if the deviations are small enough for the linearization to be valid ($|T_0 - T_w|/T_0 \ll 1$ and $|n_0^A - n_s^A|/n_0^A \ll 1$), then we only need to add a spherically symmetric term of the form $\Phi_0^{\alpha}(r, \zeta, \theta_{\zeta})$ to the right-hand side of Eq. (8), which does not contribute to the force acting on the particle. Therefore, Eq. (9) is also valid in this more general situation. Incidentally, a uniform T_w is realized when the thermal conductivity of the particle is high.

So far, we have considered the case where there is no flow of the total mixture at infinity. When there is a slow uniform flow $U_{\infty i} = (U_{\infty 1}, 0, 0)$ of the total mixture at infinity $[|U_{\infty 1}| \ll (2\kappa T_0/m^A)^{1/2}]$, we need to add $2\zeta_1 u_{\infty 1}$ and $2\hat{m}^B\zeta_1 u_{\infty 1}$ to ϕ_∞^A and ϕ_∞^B in Eq. (6), respectively, where $u_{\infty 1} = (2\kappa T_0/m^A)^{-1/2}U_{\infty 1}$. For instance, if we consider the case where the noncondensable gas is at rest, we let $u_{\infty 1} = -c_\infty \tilde{D}k/\hat{m}^B\hat{n}_0^B$. Because of the linearity of the problem, the solution of such a problem can be obtained as the sum of the solution presented in this paper and the solution of the problem of a uniform flow of the mixture past a spherical condensed phase, that is, the problem in which the same spherical condensed phase is placed in the uniform equilibrium flow of the mixture with temperature T_0 , velocity $U_{\infty i}$, and number densities of respective gases n_0^A and n_0^B . We have also analyzed this problem by the same numerical method. The drag force $F_i^D = (F_1^D, 0, 0)$ acting on the particle in this case is expressed in the following form.

$$F_1^D = 6\pi\mu L U_{\infty 1} h_D, \qquad \mu = \kappa T_0 [n_0^A / (n_0^A K^{AA} + n_0^B K^{AB}) + n_0^B / (n_0^A K^{BA} + n_0^B K^{BB})], \tag{11}$$

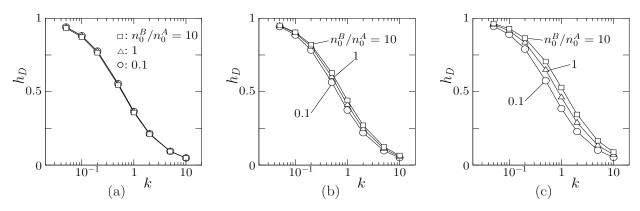


FIGURE 4. The h_D versus k in the case where $K^{AA} = K^{AB} = K^{BB}$. (a) $m^B/m^A = 0.5$, (b) $m^B/m^A = 1$, (c) $m^B/m^A = 2$.

where μ is the viscosity coefficient for the present model equation. The h^D versus k is shown in Fig. 4 for $K^{AA} = K^{AB} = K^{BB}$. In summary, the force acting on the particle when there is a uniform flow of the total mixture, in addition to the density gradient of each component considered originally in this paper, is given by $F_i^d + F_i^D$.

Finally, let us go back to the original problem without a flow of the total mixture at infinity. If the particle is left without constraint, it starts moving because of the force (9) (diffusiophoresis). The final velocity of the particle can be obtained by using the result in the preceding paragraph. That is, we superpose at infinity a uniform flow (say U_i) of the mixture that vanishes the total force ($F_i^d + F_i^D = 0$). Then the velocity of the particle relative to the flow of the total mixture ($-U_i$) gives the final velocity of the particle. From Eqs. (9) and (11), we obtain the velocity of the particle $V_i^d = (V_1^d, 0, 0)$ as

$$V_1^d = \frac{p_0 L}{6\pi} \left(\frac{2\kappa T_0}{m^A}\right)^{-1/2} \frac{D_{AB}}{\mu} \left(\frac{\partial \chi^A}{\partial X_1}\right)_{\infty} \frac{h_d}{h_D}.$$
 (12)

Following Ref. [15], we can show that the unsteadiness caused by the particle motion through the mixture with a nonuniform concentration can be neglected because of the smallness of the concentration gradient.

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